

Production of carbonate aggregates using steel slag and carbon dioxide for carbon-negative concrete



Zaid Ghouleh^a, Roderick I.L. Guthrie^b, Yixin Shao^{a,*}

^a McGill University, Civil Engineering, 817 Sherbrooke Street West, Montreal, Quebec H3A-0C3, Canada

^b McGill University, Materials Engineering, 3610 University Street, Montreal, Quebec H3A-2B2, Canada

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ABSTRACT

This work demonstrated the possibility of producing artificial carbonate aggregates from steel slag optimally activated by carbon dioxide for the purposes of waste recycling and carbon utilization in concrete. The value-added application is unique and oriented towards green concrete development. Processing of the slag involved successive carbonation treatments prior and post comminution, yielding a final CO₂-impregnated material suitably hardened and graded to replace natural aggregates in concrete. Standardized tests deemed the synthetic aggregate comparable in performance to commercial granite and Litex[®] lightweight aggregates. Compressive strength of concretes prepared using these artificial aggregates, where some specimens were further cured by carbonation, performed favorably against reference benchmark concretes. A standard full-size 20-cm concrete masonry block prepared in the manner prescribed can permanently store 2.93 kg of CO₂, outweighing its embodied carbon footprint. Extending this to annual block production in the United States and Canada, around 12 million tons of CO₂ could potentially be sequestered every year through such utilization. The final concrete products can be made carbon negative.

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1. Introduction

Concrete is the world's most used man-made material, with global production conservatively estimated at 3.8 billion cubic meters (~10 billion tons) per year [1]. It is generally a composite material composed of 10–20% Portland cement, 70–80% aggregates, and 5–10% water, by mass. Aggregates comprise the majority of the volume. The exploitation of natural resources for the acquisition of aggregates raises concerns of depletion and erosion, and faces growing environmental restrictions [2]. If more sustainable alternate sources can be suitably primed for aggregates, such as enhanced industrial wastes, concerns related to geologic preservation can be pragmatically alleviated. Recent studies have shown that carbon dioxide can considerably promote the stability, strength and physical resilience of waste steel slag [3,4], making its consideration as an aggregate replacement a promising one. In addition to preserving natural resources, concrete production could potentially be turned carbon-negative

through the use of such valorized waste derivatives, effectively turning concrete into a carbon sink.

The global production of steel is currently rated at 1.3 billion tons per year, which also amounts to the generation of approximately 400 million tons of “iron” and “steel” slag by-products [5,6]. In the United States alone, the commercial value of steel-making slag is estimated at around \$150 million [7]. Iron slag has well established uses in construction, however, steel slag bears limited recyclability. The latter originates from the downstream refinement of steel. Its limited reuse compared to iron slag is partly owed to its lack of high-temperature, suitably-hydraulic, calcium-silicate phases [8,9]. However, the main reason lies in its high free-lime (CaO) and magnesium oxide (MgO) contents, which associate deleterious expansive effects and volume instability [8,10,11]. In Canada, steel slag is banned from use in asphalt concrete and as a road-base filler [9]. Even with more stringent waste management efforts, and newly inspired potential applications, 35% of this slag is still disposed of in landfills [12].

One interesting trait of steel slag is its high specific reactivity to carbon dioxide. A number of studies have explored this phenomenon for emission abatement purposes. Huijgen et al. demonstrated the suitability of ground steel slag powder as feedstock for mineral sequestration, adding that the residue's alkalinity and chemical

* Corresponding author.

E-mail addresses: zaid.ghouleh@mail.mcgill.ca (Z. Ghouleh), roderick.guthrie@mcgill.ca (R.I.L. Guthrie), yixin.shao@mcgill.ca (Y. Shao).

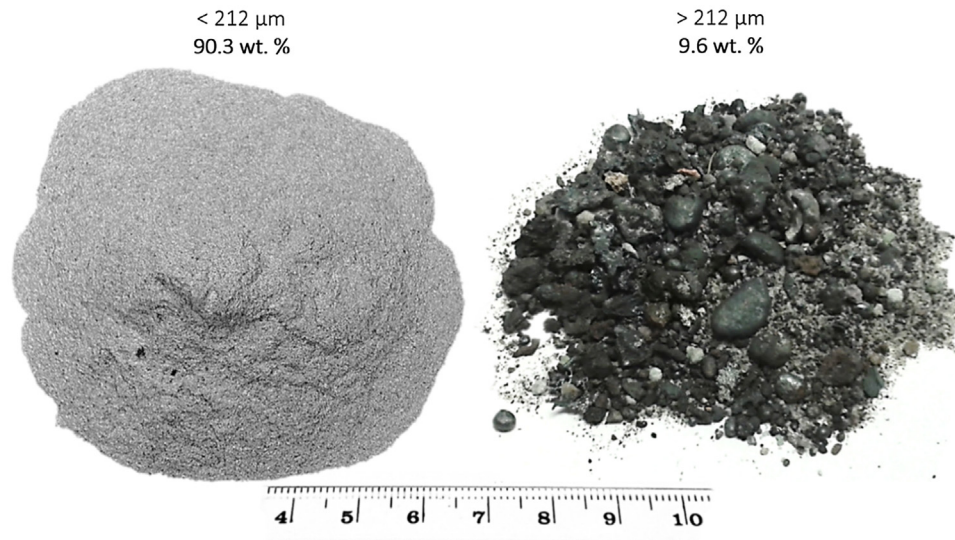
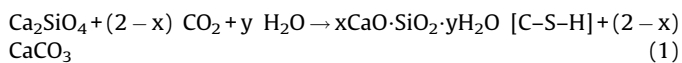


Fig. 1. Sieve separated as-received raw KOBM steel slag, left, slag passing 212 μm sieve; right, metallic impurities retained on the 212 μm sieve.

instability make it more reactive than virgin minerals [13]. Zevenhoven et al. suggest that steel industry slag has the current capacity to fixate 60–80 million tons of CO₂ per year globally [14]. Other works target the generation of precipitated calcium carbonate (PCC) and/or ground calcium carbonate (GCC) products from slag to serve as filler materials, most commonly used by the papermaking industry [15].

Steel slag is particularly rich in di-calcium silicates (C₂S) and can rapidly react with CO₂ to form calcium–silicate–hydrate (C–S–H) and CaCO₃, as generically depicted in Eq. (1). C–S–H is a phase found abundantly in hardened concrete and formed during the hydration of cement. It contributes to strength and other physico-mechanical developments within concrete, and is largely responsible for the binding properties of hydrated cement [16]. The generation of C–S–H from steel slag via carbonation therefore presents promising valorizing options for binder applications.



Moreover, carbonation can rid the slag of its expansive behavior by consuming free-lime content [13], as per the overall reaction in Eq. (2). It also promotes the slag's strength through C–S–H formation and carbonate precipitation [17,18], qualifying the slag for previously restricted applications. Heavy metal mobility can be hindered by carbonation's pH neutralizing effect, with one study reporting reductions of up to 80% in heavy metal leaching [19]. A thorough understanding of the mechanism of carbonation can therefore be exploited to impart various improvements in steel slag. Considering the abundance of waste slag, the lack of its complete reuse leaves considerable room for improvement.



In this paper, the valorizing potential of carbonation was exploited to process steel slag from a local mill into an artificial CO₂-laden carbonate aggregate material to replace natural stone and maximize the carbon utilization potential in concrete. The synthesis of fine aggregates, $\leq 5\text{ mm}$, was targeted, as this size fraction bears more prevalent effects on the properties of concrete than coarser gradations; this fraction's shape and particle size distribution profoundly influence concrete's strength and overall resilience [20,21]. Unlike mineral sequestration, CO₂ was not just simply stored in concrete, it was in fact utilized here to activate the superior strength and stability of otherwise landfilled waste steel

slag. The implications of this work seeks to alleviate ecologically-taxing effects inflicted simultaneously by the steel and concrete industries, both of which are energy, emission, and resource intensive. The presented demonstration offers a novel and sustainable construction practice with merits for waste recycling, resources conservation, and CO₂ utilization and sequestration.

2. Materials and methods

2.1. Steel slag

The steel slag used in this study was obtained from the RioTinto Iron & Titanium (RTFT) operation located in Sorel-Tracy, Quebec, Canada. It originated from the plant's Klockner Oxygen Blown Maxhutte (KOBM) process, a technically improved variant of the more conventional Basic Oxygen Furnace (BOF). The as-received slag was passed through a 212 μm sieve to remove large metallic fragments and other impurities to ensure homogeneity. The sieve-separated slag and metallic impurities are shown in Fig. 1. It was found that the bulk of the iron content was confined to particles with diameters between 0.3 and 10 mm, hence the effectiveness of sieve-separation. Table 1 presents the composition of the slag portion passing the 212 μm sieve as obtained by X-Ray Fluorescence (XRF) and Carbon/Sulfur (C/S) analysis. Note the low iron content and uncharacteristically high CaO as a result of excluding the $>212\ \mu\text{m}$ portion.

Table 1
Chemical composition of KOBM steel slag.

Composition wt% (DL=0.01%)			
Oxide	Weight%	Oxide	Weight%
SiO ₂	24.92 ± 1.34	Na ₂ O	<DL
TiO ₂	0.50 ± 0.03	K ₂ O	<DL
Al ₂ O ₃	1.83 ± 0.21	P ₂ O ₅	0.04 ± 0.01
Fe ₂ O ₃	3.04 ± 1.19	Cr ₂ O ₃	0.05 ± 0.01
MnO	0.32 ± 0.03	V ₂ O ₅	0.03 ± 0.01
MgO	4.89 ± 0.61	Total-C	0.14 ± 0.01
CaO	61.21 ± 0.40	Total-S	0.53 ± 0.04
LOI	<DL		

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